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Radioactive Species Produced by Cosmic Rays in Bruderheim and Other Stone Meteorites

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Abstract. In this paper we report measurements of the activity of 16 radioactive nuclides in the chondritic meteorite Bruderheim, which fell on March 4, 1960. The data are compared with those for the iron Aroos and other stone, iron, and stony-iron meteorites. For nuclides of $A \geqq 46$, the activities normalized to iron and nickel content are closely similar. For lower A, the activities in stone are mainly produced in lighter elements. The spectrum of bombarding particles is shown to be closely similar in irons and stones. The results are consistent with a constant cosmic-ray intensity. Cosmic-ray bombardment ages of 30×10^6 years for Bruderheim and 130×10^6 years for Admire are calculated from rare-gas data and the activities of Na^{22}, Al^{26}, and Cl^{36}.

The record of cosmic-ray bombardment in meteorites is in the form of rare stable and radioactive nuclei produced by transmutation. Many measurements of the concentration of such nuclei have been made, and they have been used to draw conclusions about both cosmic rays and meteorites. The field has recently been reviewed [*Arnold*, 1961].

Honda, Shedlovsky, and Arnold [1961] have reported the activity of several long-lived radioactive nuclides in four iron meteorites, and *Honda and Arnold* [1961] have measured a large number of short- and long-lived species in the freshly fallen iron Aroos. From these data and those of other workers it has been concluded [*Arnold, Honda, and Lal,* 1961] that the cosmic-ray intensity, averaged over the half-life of each species, has been constant within the limits of error of theory and experiment. It has been possible to account with adequate precision for the production rates and concentrations of radioactive and stable species in iron meteorites. Some predictions of this model have been verified [*Stauffer and Honda,* 1961].

It is also of interest, as part of the same program, to measure the concentrations of as many nuclides as possible in stony meteorites [*Ehmann and Kohman,* 1958]. This paper is mainly concerned with the measurement of a number of radioactive species in the recently fallen chondrite Bruderheim. We also report here earlier measurements of long-lived activities in the chondrite Achilles, and in the separated stone and iron fractions of the pallasites Admire and Brenham. A description of these meteorites is given in the Appendix. We will discuss the meaning of these results for the energy distribution of the bombarding particles, the constancy of the cosmic-ray intensity in time and space, and the cosmic-ray bombardment ages of these meteorites.

The Bruderheim chondrite fell on March 4, 1960, near Edmonton, Alberta, Canada. The fall consisted of many stones; the total weight exceeded 100 kg. This is a typical gray chondrite whose composition is very close to the mean of the low-iron group of *Urey and Craig* [1953] [*Baadsgaard, Campbell, Cumming, and Folinsbee,* 1961; *Duke, Maynes, and Brown,* 1961]. Through the courtesy of Dr. Folinsbee, we received a large specimen on May 9, 1960.

Using this sample we have measured the content of 16 radioactive nuclides, including most of those reported for Aroos and Fe^{55} (an upper limit was also set for Be^7). Wet chemical procedures were used throughout. Isolation steps and counting methods were similar to those for Aroos. The activities of species of $A \geq 46$, for a given weight of Fe, Co, or Ni, are close to those found in Aroos, except for special cases such as the neutron capture species Co^{60}. The measurements of long-lived Cl^{36}, Al^{26}, and Be^{10} agree with those for the other stones, when chemical composition is taken into account.

These data, together with the rare-gas concentrations, yield bombardment ages of 30 ×

TABLE 1. Content of Cosmic-Ray-Produced Radioactivities in Bruderheim*

Nuclide	$t_{1/2}$	dpm/kg at Time of Fall
Be^7	53 d	≤ 100
Be^{10}	2.5×10^6 y	19 ± 2
Na^{22}	2.58 y	90 ± 10
Al^{26}	7.4×10^5 y	60 ± 6
Cl^{36}	3.1×10^5 y	7.5 ± 0.8
Sc^{46}	83.8 d	6.2 ± 0.6
Ti^{44}	~ 200 y	2.0 ± 0.2
V^{48}	16.0 d	34 ± 7
V^{49}	330 d	34 ± 5
Cr^{51}	27.8 d	110 ± 27
Mn^{53}	$\geq 2 \times 10^6$ y	85 ± 17
Mn^{54}	308 d	100 ± 13
Fe^{55}	2.6 y	340 ± 80
Co^{56+58}	~ 74 d	14 ± 4
Co^{57}	240 d	11 ± 1
Co^{60}	5.26 y	9 ± 1
Ni^{59}	8×10^4 y	12 ± 3

* Other data: Ar^{39} 10 ± 1; Ar^{37} 23 ± 4; T 260 ± 30 [*Fireman and DeFelice*, 1961].

10^6 years for Bruderheim and 130×10^6 years for Admire.

Experimental Procedures

Chemical treatment. A total of 850 grams of Bruderheim was dissolved in HF and H_2SO_4. After drying, the cake was extracted with water. The residue was fused with NaOH and extracted with HCl; Cr and other elements were recovered from this fraction (and later combined with the proper fractions from the main solution). Carriers of Sc, V, and Be were added to the main solution. Fe was separated by ether extraction. A large precipitate of Mg and Al was removed from saturated HCl solution. We note that throughout the process the removal of Mg without excessive losses of other elements was a major difficulty. Hydroxides were precipitated with NH_4OH, leaving most Ni in solution. A second $MgCl_2$ precipitation was carried out from saturated HCl. It was necessary later to treat the chloride precipitates to recover various hydroxide group elements. The solution was neutralized again with NH_4OH, and the precipitate was dissolved in 1 *M* HCl and H_2O_2. The cations were separated on a 1-liter cation-exchange column, eluting with 1–6 *M* HCl. The order of recovery was V, Ti, Be, Mn, Al, Cr, and Sc. A sulfide precipitation removed Ni and Co from the NH_4OH solutions. Na was recovered from the supernate by cation exchange, after treatment with $Ba(OH)_2$.

For Cl a separate sample of 80 grams was fused with NaOH. After extraction the residue was attacked with dilute HNO_3. Chloride carrier was added to the combined solutions, and AgCl precipitated.

The Achilles meteorite (100 grams) was attacked with HNO_3 and HF at room temperature. The Cl fraction was recovered from the filtrate as AgCl. The residue from the acid treatment was combined with the filtrate and treated further at high temperature with H_2SO_4. The water extract was saturated with HCl gas at low temperature, and a white precipitate containing Al was separated. Fe^{III} was removed by anion exchange. After addition of excess NaOH and Be carrier, Be and Al remained in the filtrate.

The stone phase of Admire (75 grams) was treated with HF and H_2SO_4. Fe^{III} was removed by ether extraction in HCl solution. Al was precipitated using HCl-ether. Be was recovered from the filtrate.

The metal phases of Admire (340 grams) and Brenham (200 grams) meteorites were treated by a procedure similar to those reported previously [*Honda, Shedlovsky, and Arnold,* 1961]. They were dissolved in nitric acid, and Cl^{36}, Be^{10}, Al^{26}, K^{40}, Mn^{53}, and Ni^{59} were separated.

Counting methods. Our counting methods, for β, γ, and X radiation, are described in earlier papers [*Honda and Arnold,* 1961]. The counting samples were usually more massive than those from Aroos. The self-absorption factor is especially important for Na^{22}, Al^{26}, Cr^{51}, and Co.* For Fe^{55} and Ni^{59} the samples were nearly 'infinitely thick.' For Na^{22} and Al^{26} in Bruder-

TABLE 2. Content of Cosmic-Ray-Produced Radioactivity in Some Stone and Stony-Iron Meteorites, dpm/kg

	Achilles	Admire Stone	Admire Metal	Brenham Metal
Be^{10}		14 ± 2	1.7 ± 0.3	0.2 ± 0.4
Al^{26}	50 ± 5	43 ± 4	1.5 ± 0.5	0.1 ± 0.5
Cl^{36}	6.0 ± 0.6		7.4 ± 0.9	0.1 ± 0.2
K^{40}			1.1 ± 0.4	
Mn^{53}			200 ± 20	<15
Ni^{59}			300 ± 30	<20

heim, positrons were counted in both β and γ counters.

The nuclides were identified by (1) recycling to constant specific activity; (2) absorption measurements for Be^{10}, Na^{22}, Sc^{46}, and Co*; (3) decay measurements for Sc^{46}, V^{48}, V^{49}, Cr^{51}, Mn^{54}, and Co*.

Results and Discussion

The results for Bruderheim are shown in Table 1; the others, in Table 2. The accuracy of the data is calculated from the standard deviation of counting statistics. Ten per cent is taken as the minimum, including other sources of error [*Honda and Arnold*, 1961].

Because of the shorter time between fall and measurement as well as the greater size of the sample, the V^{48} and Cr^{51} data for Bruderheim are better than those for Aroos. The figure for Be^7 is still only an upper limit. The chemical yield for each element, except Sc, V, and Be, is calculated from the analytical data. We have assumed concentrations of 10 ppm Sc, 30 ppm V, and negligible Be, in addition to added carrier.

Table 2 shows the results obtained in other meteorites. Within experimental error no activity is present in our specimen of Brenham metal. Signer (private communication) reports the absence of cosmogenic rare gases. Since helium has been measured in another specimen of this meteorite, we must conclude that our sample was buried deeply within the original mass. The activities for Admire metal are about half those in Aroos, except for the high value of Ni^{59}. The activities in Achilles and Admire stone compare reasonably (see below) with those in Bruderheim. Our Al^{26} measurements are consistent with the results reported by *Ehmann and Kohman* [1958]. The Al^{26} content of chondrites has also been checked by other workers using different methods [*Van Dilla, Arnold, and Anderson*, 1960]. For Be^{10}, however, much lower values were reported by Ehmann and Kohman: 1.6 ± 0.1 and 5.1 ± 0.5 dpm/kg in Plainview and Richardton, respectively. Their values are comparable to those obtained for small iron meteorites. Because of the high abundance of oxygen, the production rate in chondrites must be substantially higher. Their low figures might possibly be due to incomplete separation of Be from Al.

Vilcsek and Wänke [1960] reported the Na^{22} content of the chondrite Breitscheid (fell in 1956). Their figure, 89 ± 15 dpm/kg, is in good agreement with ours. The same authors [1961] tried to measure Cl^{36} in Bruderheim by extraction with dilute acid from a powdered sample. They obtained 5.7 ± 0.4 dpm/kg, which is close to our value even before considering possible incompleteness of the extraction.

Rowe and van Dilla [1961] report values of 60 dpm/kg Al^{26}, 90 dpm/kg Na^{22}, and 82 dpm/kg Mn^{54} in Bruderheim. These values, obtained by γ-ray spectrometry on an intact specimen, are in excellent agreement.

Comparison with Aroos and other iron meteorites. The activities measured in Bruderheim can be compared with those of a small iron meteorite such as Aroos. In Table 3 we have multiplied the activities found in Aroos [*Honda and Arnold*, 1961] by a factor of 0.24 (0.18 for Ni and Co nuclides). The factor 0.24 corresponds to the sum of the Fe, Co, and Ni concentrations in Bruderheim in weight per cent, whereas 0.18 is

TABLE 3. Content of Radioactivity in Bruderheim and Aroos Meteorites at Time of Fall, dpm/kg

Nuclide	Bruderheim	0.24 × Aroos	Ratio
Be^{10}	19	1	19 ± 2.7
Na^{22}	90	0.4	200 ± 30
Al^{26}	60	0.9	70 ± 12
Cl^{36}	7.5	3.3	2.3 ± 0.3
Ar^{37}	23	5*	4.6 ± 0.6
Ar^{39}	10	4*	2.5 ± 0.4
Sc^{46}	6.2	7.2	0.86 ± 0.12
Ti^{44}	2.0	1.1	1.8 ± 0.3
V^{48}	34	22	1.5 ± 0.7
V^{49}	34	38	0.89 ± 0.16
Cr^{51}	110	65	1.7 ± 0.8
Mn^{53}	85	124	0.69 ± 0.10
Mn^{54}	100	113	0.89 ± 0.12
Fe^{55}	340	380†	0.89 ± 0.45
		0.18 × Aroos	
Co^{56+58}	14	22	0.64 ± 0.20
Co^{57}	11	16	0.69 ± 0.10
Co^{60}	9	3	3 ± 0.4
Ni^{59}	12	11	1.1 ± 0.3

* From *Fireman and DeFelice* [1960].

† The value of 1600 ± 600 dpm/kg is due to M. Honda (unpublished).

TABLE 4. Comparison of Specific Activities in Stone and Metal

Stone				Metal		
dpm/kg Target	Bruderheim	Achilles	Admire Stone	dpm/kg Target	Admire Metal	Aroos
Be^{10}/O	53 ± 5		32 ± 4	$Be^{10}/Fe + Ni$	1.7 ± 0 3	4.1 ± 0.4
$Al^{26}/Si + Al$	300 ± 30	250 ± 25	220 ± 22	$Al^{26}/Fe + Ni$	1.5 ± 0.5	3.6 ± 0.4
$Cl^{36}/Ca + K$*	320 ± 64	210 ± 40		$Cl^{36}/Fe + Ni$	7.4 ± 0.9	14 ± 1.4
$Mn^{53}/Fe + Ni$	420 ± 100			$Mn^{53}/Fe + Ni$	200 ± 20	515 ± 52
Relative bombardment intensity	0.82	0.65	0.55		0.45	1.00

* Corrected for contribution from target Fe + Ni.

the relative content of Co and Ni. The ratios are not far from unity between Sc^{46} and Ni^{59} except for Co^{60} and Ti^{44}. The weighted average (excluding these two nuclides) is 0.82. There is no trend with ΔA (defined as $A_{target} - A_{product}$). We conclude that the relative spectrum of nuclear-active particles [*Arnold, Honda, and Lal,* 1961] in small iron meteorites and in Bruderheim is effectively the same for the production of species from Sc^{46} to Ni^{59} and that the flux in our Bruderheim sample was about 80 per cent of that in our sample of Aroos. The excess of Co^{60} we attribute to a higher slow neutron flux in Bruderheim. This is to be expected, since neutrons are more effectively moderated by low-*Z* elements. The activity of Co^{60} in Bruderheim is about 10^4 dpm/kg Co. The small excess of Ti^{44} may be ascribed to production from Ti and Cr.

For the other nuclides in this group, neutron capture and production from higher elements are not expected to be important.

From Be^{10} to Ar^{39}, the large differences observed must be attributed to production from other major constituents closer to the product nuclides.

Comparison of production in iron and stone phases. In Table 4 we compare the activities of some long-lived isotopes in Bruderheim, Achilles, the two phases of Admire, and Aroos. The activities have been divided by the weight per cent of the target elements which are most important for each isotope. This corrects (accurately enough for present purposes) for the effect of changes of composition. The last row gives the relative bombardment intensities in the various samples as determined from activity ratios. The value of 0.82 for our sample of Bruderheim is taken from the previous section.

The most important conclusion from these data and those in Table 3 is that the spectrum of bombarding particles in Bruderheim or Admire is closely similar to that in Aroos, a typical small iron meteorite. One direct demonstration of this is the lack of a trend in the ratio of the observed activities in Bruderheim and Aroos in going from Sc^{46} to Fe^{55}. The former is produced mainly at energies of several hundred Mev, the latter mainly in the region of 10 to 20 Mev. A relative change of 40 per cent in the two differential spectra over the range from 20 to 500 Mev [*Arnold, Honda, and Lal,* 1961] would have produced a visible trend.

The comparison of the bombardment intensities in the two phases of Admire allows us to extend this result to higher energies. The species Be^{10} and Al^{26} are produced in iron mainly in the bev region. In stone they are produced mainly in O and Si respectively, by reactions that have large cross sections at low energies. They are, therefore, like Mn^{54} in iron, produced mainly by low-energy particles. The relative bombardment intensities of 0.55 and 0.45 in the two phases are nearly the same. If the difference is significant, it can be accounted for by a difference of about 20 per cent in the spectra in our specimens of Admire and Aroos between low energies and the bev region. The difference would be in the direction of a 'softer' spectrum in Admire. The lower bombardment intensity in our specimen of Admire is consistent with a small effect in this direction.

Bruderheim and Achilles are low-iron chondrites. We may expect the relative production

TABLE 5(*a*). Cosmic-Ray-Produced Rare-Gas Content: Unit: 10^{-8} cc/g (NTP)

	Bruderheim*	Admire Stone*	Admire Metal†	Brenham Stone*	Brenham Metal†
Ar^{38}	1.22‡	0.49	3.9	0	≤0.03
Ar^{36}	0.80‡	0.34	2.5	0	
Ne^{22}	9.6	41	0.76		≤0.01
He^3	43.5	101	63	0.2	≤0.5

TABLE 5(*b*). Bombardment Age: Unit: 10^6 years

	Bruderheim	Admire Stone	Admire Metal
Cl^{36}–Ar^{36}	33 ± 8		140 ± 20
Na^{22}–Ne^{22}	27 ± 4		
Al^{26}–Ne^{22}		120 ± 20	
Average	30 ± 4	130 ± 14	

* H. Stauffer (private communication). Estimated errors: ±10–15 per cent for Ne and He, ±20 per cent for Ar.

† *Signer and Nier* [1961].

‡ The data on the Ar isotopes have been corrected for primordial gas, using the procedure described by *Stauffer* [1961]. The uncorrected values were Ar^{38}, 1.33; Ar^{36}, 1.39.

rates of all species to be similar in other representatives of this class. The production in stones or stone phases of substantially different composition may be estimated from the data in Tables 3 and 4. We note that an appreciable fraction of Cl^{36} and the Ar isotopes is produced from Fe, in addition to that made from Ca + K.

Calculation of production rates. In another paper [*Arnold, Honda, and Lal,* 1961] we have described a method of calculating production rates of various species, using cross sections along with experimental data of cosmic-ray physics. The resulting relative and absolute production rates are in good agreement with the data for Aroos and other iron meteorites. Since it has been demonstrated above that the relative differential spectra of the bombarding particles in Bruderheim and Admire were closely similar to that in Aroos, the same method is also applicable in these cases. However, the necessary cross-section data for the targets O, Mg, Si, Ca, and others are not yet available, especially at the low energies that are most important.

The consistency of the production rates may be seen from the data on Bruderheim in Table 4. The production rates of Ar^{37} and Ar^{39} are discussed by *Stoenner, Schaeffer, and Davis* [1960] and *Fireman and DeFelice* [1961]. The production rates of Al^{26} and Cl^{36}, normalized to the weight of the main target elements, are similar to those of Mn^{54} and Mn^{53}, species of similar ΔA produced in Fe. The ratio Na^{22}/Mg + Na amounts to 570 dpm/kg; since a significant contribution from Si and Al must be present this value is also close to the others. The value of Be^{10}/O is much lower than that for the comparable case of V^{49}/Fe, as is to be expected, since the cross section for Be^{10} production by 220-Mev protons in a CNO target is quite low, in fact several times lower than that for Be^7, according to unpublished data of Honda and Lal. Thus the production rates of the species produced in light elements appear to be consistent with the others.

The data on Aroos and other iron meteorites led us to the conclusion that the cosmic-ray intensity, averaged over the half-life of each radioactive product nuclide, has been nearly constant. The data on Bruderheim and other stones also appear, from this discussion, to be consistent with the conclusion that secular equilibrium prevails.

Cosmic-ray-produced isotopes of heavier elements. The production of Fe^{55} by the $(n, 2n)$ reaction reaches almost 2000 dpm/kg, or 2 dpm/g target. The product of a low-energy reaction such as (n, p), $(n, 2n)$, or (n, pn) on another element might possibly be made at a rate as high as 10 atoms/g min. The activity of the (n, γ) product Co^{60} reaches 10 atoms/g min in Bruderheim; still higher values are possible, of course, if the capture cross section is very large. Over a bombardment period of 10^8 years, a rate of 10 atoms/g min would result in the transmutation of about 4×10^{-8} of the target atoms. This might in special cases give a detectable result even for trace-element targets.

Rare-gas content and cosmic-ray age. A comparison of the activity data with the concentrations of stable rare gases permits the calculation of cosmic-ray ages. Rare-gas measurements on our samples were made by P. Signer and H. Stauffer (unpublished). The data are given in

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Table 5(*a*). The calculated bombardment ages are shown in Table 5(*b*).

For Bruderheim, we have used the pairs Na^{22}-Ne^{22} and Cl^{36}-Ar^{36}. We have assumed 1:1 for the direct production ratio Na^{22}:Ne^{22}. For Cl^{36}:Ar^{36}, we have assumed 1:1 for production from Ca + K, and the usual value of 4:1 for production from Fe. From these we calculate a ratio of 6:4. For Cl^{36}:Ar^{36} in Admire metal, the usual ratio of 4:1 has been taken. The Al^{26}-Ne^{22} age in the stone phase has been calculated as follows. We estimate that the ratio Al^{26}:Na^{22} is one-third lower in the Admire stone phase than in Bruderheim, because the Si:Mg ratio in Admire stone is only about half as great. On this basis the Na^{22} content of Admire stone at the time of fall was the same as that of Bruderheim, and the age therefore four times greater.

The values in Table 5 are mutually consistent. Incorrect assumptions about production ratios, and possible diffusion losses, are possible sources of error. *Vilcsek and Wänke* [1960] have obtained an age of 30 × 10^6 years for the chondrite Breitscheid, using the same assumptions for the pair Na^{22}-Ne^{22}. Values around 20 × 10^6 years or less have been obtained for other chondrites by the H^3:He^3 method [*Geiss, Hirt, and Oeschger*, 1960]. The data are not sufficient to permit a useful discussion of this difference.

Appendix. Meteorite Samples

Admire. Stony-iron meteorite, Brecciated pallasite. Lyon County, Kansas, USA. Found 1881. Total mass collected more than 50 kg. Our specimen was a composite of Ward Catalog no. S 1330 (271 g); S 1327 (337 g); S 1335 (312 g): Measured density 4.55 (S 1335).

Brenham. Stony-iron meteorite, pallasite. Brenham Township, Kiowa County, Kansas, USA. Found 1882. Total mass collected several tons. Our specimen: Ward Catalog no. 10:50 (365 g); no. 10:54 (259 g). Measured density 4.88–4.98 (corresponding to a 1:1 weight ratio of stone and metal phases).

Achilles. Stone meteorite, veined crystalline chondrite. Rawlins County, Kansas, USA. Found 1924. Stone. Ward Catalog no. S 1826 (445 g).

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